

IIIIII Microgravity Experiments by Aircraft Parabolic Flights I IIIII
(Original Article)

Measurement of Surface Tension of Molten Metal under Reducing Gas Atmosphere in Microgravity Condition during Parabolic Flight — Attempt to Control Oxygen Partial Pressure —

Shumpei OZAWA ¹, Noriaki TAKENAGA ², Takashi INOUE ¹, Yuto TAKEI ¹,
Taketoshi HIBIYA ³, Akitoshi MIZUNO ⁴ and Masahito WATANABE ⁴

Abstract

In order to establish technology for precise measurement of surface tension of molten metals under microgravity condition during the parabolic flight of an airplane, copper was selected as the sample from the viewpoints of its oxidization tendency, oxygen partial pressure dependence of surface tension of the molten sample, and ease of heating with electromagnetic levitation. The non-combustibility of the Ar-3vol. %H₂ gas that can reduce the oxygen partial pressure of the measurement atmosphere was experimentally confirmed as stipulated in the civil aeronautics law in order to employ it in the parabolic flight experiment. The surface tension of molten copper was successfully measured under the reducing gas atmosphere in microgravity during a parabolic flight experiment for the first time.

Keyword(s): Surface tension, electromagnetic levitation, oxygen partial pressure, parabolic flight, microgravity

1. Introduction

Various high-value-added high-temperature melt processes such as the precision casting of heat resistant turbine blades and the crystal growth of large-diameter semiconductor silicon have become more and more complicated and diversified in recent years to respond to difficult demands for the further improvement of products and manufacturing, e.g. an improvement in the quality and performance of products, cost reductions, energy saving, and a reduction of product manufacturing time. Thus it is indispensable to employ a numerical calculation together with an experiment to optimize the processes. In order to obtain reliable numerical calculation results, accurate thermophysical properties are required. In particular, an accurate surface tension and its temperature coefficient are very important if the process has a free melt surface at which the Marangoni convection appears due to the surface tension gradient.

The oscillating droplet technique using electromagnetic levitation (EML) is one of the most promising methods to measure the accurate surface tension of molten metals¹⁻³. With this technique, metallic sample can be levitated by the Lorentz force generated from the interaction between an electromagnetic field and a loop current at the sample surface induced by the radiofrequency current which flows through the levitation coil. The sample is melted by Joule heating in containerless state.

The major advantages of this technique are to assure measurements free of contaminants from the container, at temperatures sufficiently above the melting point of the sample, and under highly undercooled conditions. This is attributed to the sample not being in contact with the container, which is chemically reactive with the sample at such high temperatures and plays a role in the nucleation center below the melting point. Furthermore, this technique makes it easy to control the oxygen partial pressure (P_{O_2}) of atmospheric gas, which is an important factor influencing the surface tension of molten metals

When only the surface tension acts as a restoring force in a levitated droplet without any external force such as gravity, it can be calculated from the frequency of surface oscillation and sample mass using the following the Rayleigh equation⁴,

$$\sigma = \frac{3}{8} \pi M \nu_R^2 \quad (1)$$

where σ is the surface tension, M is the sample mass, and ν_R is the single surface oscillation of $l = 2$ mode known as the Rayleigh oscillation.

On the ground, the frequency of the $l = 2$ mode splits into three, corresponding to oscillations of $m = 0, \pm 1$, and ± 2 ⁵ because of the droplet deformation by gravitational acceleration and electromagnetic forces from the levitation coil. As a result, the effect of the droplet deformation must be calibrated by theoretically derived Cummings and Blackburn equation⁵.

1 Department of Mechanical Science, Chiba Institute of Technology, 2-17-1 Tsudanuma, Narashino, Chiba 275-0016, Japan

2 Department of Aerospace engineering, Tokyo Metropolitan University, 6-6 Asahigaoka, Hino, Tokyo, Japan (Present affiliation: Ricoh Co., Ltd)

3 System Design and Management Research Institute, Keio University, 4-1-1, Hiyoshi, Kohoku-ku, Yokohama, Kanagawa 223-8526, Japan

4 Department of Physics, Gakushuin University, 1-5-1 Mejiro, Toshimaku, Tokyo 171-8588, Japan

(E-mail: shumpei.ozawa@it-chiba.ac.jp)

$$v_R^2 = v_{2,0}^2 - v_l^2 \{3.832 - 0.1714(Z_0/a)^2\} \quad (2)$$

$$v_R^2 = v_{2,\pm 1}^2 - v_l^2 \{3.775 + 0.5143(Z_0/a)^2\} \quad (3)$$

$$v_R^2 = v_{2,\pm 2}^2 - v_l^2 \{-0.9297 + 2.571(Z_0/a)^2\} \quad (4)$$

$$a = \sqrt[3]{\frac{3M}{4\rho\pi}} \quad (5)$$

$$Z_0 = \frac{g}{8\pi^2 v_l^2} \quad (6)$$

$$v_{l,0}^2 = \frac{1}{3} \sum_{m=1}^l v_{l,m}^2 \quad (7)$$

where, $v_{2,0}$, $v_{2,\pm 1}$, and $v_{2,\pm 2}$, are the frequencies of the $m = 0, \pm 1$, and ± 2 oscillations for the $l = 2$ mode, v_l is the frequency of motion of the center of gravity for the droplet, a is the radius when the sample becomes a sphere, ρ is the density of the sample, and g is the gravitational acceleration. In order to prove the Cummings and Blackburn equation experimentally, several attempts were made to measure the surface tension of molten metals using the EML under microgravity condition. However it has not yet been validated completely because P_{O_2} of the atmospheric gas was not controlled and measured in these studies⁽⁶⁻⁸⁾ and P_{O_2} strongly influences the surface tension of molten metals.

In this paper, we selected copper as the sample for the surface tension measurement of molten metals under microgravity condition during the parabolic flight of an airplane in the light of the influence of oxygen partial pressure of atmospheric gas on the experiment. Furthermore, the results of terrestrial experiment stipulated in the Japanese Civil Aeronautics Act described that Ar-3vol.%H₂ gas can be employed in measurements in parabolic flight experiments, even though it contains combustible H₂ gas. The purpose of this study was to establish technology for precise measurement of surface tension of molten metals under microgravity condition during the parabolic flight of an airplane.

2. Selection of experimental sample and P_{O_2}

In order to measure the precise surface tension of molten metals using EML under microgravity during the parabolic flight experiment, it is necessary to clear several limiting conditions. When small Gulfstream-II jet plane operated by Diamond Air Service Inc (Aichi, Japan) is used for the parabolic flight experiment, the roughly 20 seconds of microgravity condition is preceded by a hypergravity condition from 1.5 G to 2.0 G for 20 seconds in order to accelerate the airplane⁽⁹⁾. It is favorable that the sample can be levitated even under this gravity variation to offer greater flexibility to the experiment profile in this case. It is important for the sample to be melted

within a short time. Furthermore, the melt should not be oxidized even at comparatively high P_{O_2} because only a limited type of gas is acceptable for use in an airplane from the perspective of safety. This is also essential for the viscosity measurement of the droplet under the microgravity condition. In addition to these, it is favorable that the influences of temperature and oxygen partial pressure on surface tension of the candidate sample have been reported even if in only a few reports.

For these reasons, copper was selected as the sample for surface tension measurement under microgravity condition during the parabolic flight experiment. **Figure 1** shows the temperature dependence of the equilibrium P_{O_2} of copper oxide, CuO and Cu₂O, which were calculated from the following Gibbs energy for formation of these oxides⁽¹⁰⁾.

$$\Delta G_{CuO(s)}^\circ = -306220 + 173.30T \text{ (J} \cdot \text{mol}^{-1}) [298-1357 \text{ K}] \quad (8)$$

$$\Delta G_{Cu_2O(s)}^\circ = -338300 + 146.63T \text{ (J} \cdot \text{mol}^{-1}) [298-1357 \text{ K}] \quad (9)$$

$$\Delta G_{Cu_2O(s)}^\circ = -380110 + 177.35T \text{ (J} \cdot \text{mol}^{-1}) [1357-1517 \text{ K}] \quad (10)$$

$$\Delta G_{Cu_2O(l)}^\circ = -243390 + 87.34T \text{ (J} \cdot \text{mol}^{-1}) [1517-2000 \text{ K}] \quad (11)$$

where T is the temperature. Since a levitated metal droplet usually shows a large undercooling, we assumed that the surface tension of molten copper is measured from an undercooled condition above 1300K, at which the undercooling level is only 58K. In this case, the melt is not oxidized even under comparatively high P_{O_2} of 10⁻¹ Pa according to **Fig.1**.

Figure 2 exhibits the literature data of the influence of P_{O_2} on the surface tension of molten copper at 1373 K and 1383 K⁽¹¹⁻¹⁵⁾. Since a conventional surface tension measurement technique such as the sessile drop method can assure the measurement at a comparatively low temperature to prevent the chemical reaction between the sample and measurement device, high temperature data has not been reported apart from these. In these measurements, the P_{O_2} was mainly controlled by the following gas phase equilibria for the formation of H₂O and CO₂.



For comparison, we measured the surface tension of molten copper under Ar-He-3vol.%H₂ and Ar-He-10vol.%H₂ gases using the EML on the ground in this study. The P_{O_2} of the H₂-containing gas varies depending on temperature as in the case of CO-containing gas because the following equilibrium constant for formation of H₂O described in the reaction (12), K_{H_2O} , has a temperature dependence.

$$K_{H_2O} = \frac{P_{H_2O}}{P_{H_2} \cdot \sqrt{P_{O_2}}} \quad (14)$$

where $P_{\text{H}_2\text{O}}$, P_{H_2} and P_{O_2} are the partial pressures of gases for H_2O , H_2 , and O_2 . The temperature dependences of P_{O_2} for these gases were evaluated using the following standard Gibbs energy of formation of H_2O^{16} , in which the $K_{\text{H}_2\text{O}}$ was calculated from the P_{O_2} measured by zirconia oxygen sensor operated at 1008K.

$$\Delta G_{\text{H}_2\text{O}}^{\circ} = -246535 + 54.94T \text{ (J} \cdot \text{mol}^{-1}\text{)} \quad (15)$$

The P_{O_2} measured by the oxygen sensor indicated about 10^{-23} Pa and 10^{-24} Pa for gases of Ar-He-3vol. % H_2 and Ar-He-10vol.% H_2 , respectively. The ratio between the $P_{\text{H}_2\text{O}}$ and P_{H_2} was assumed to be constant regardless of temperature in evaluation P_{O_2} . This figure confirms that the surface tension of molten copper is decreased with increasing P_{O_2} . Our measurement results well agree with the literature data. From the results of **Fig. 1** and **2**, it is essential to clarify the P_{O_2} during the measurement of the surface tension of molten copper. Moreover, it should be less than 10^{-1} Pa. However, even if high purity copper is employed in the measurement, it is very

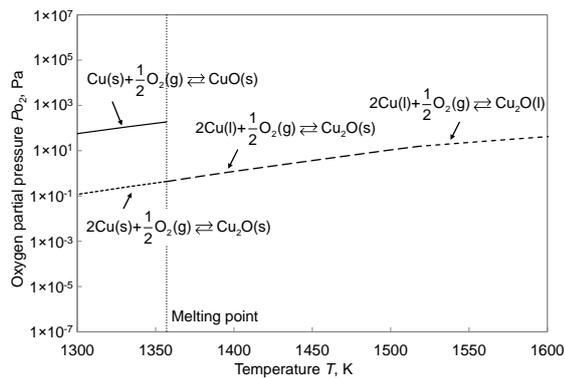


Fig. 1 Equilibrium oxygen partial pressure for formations CuO and Cu_2O as a function of temperature

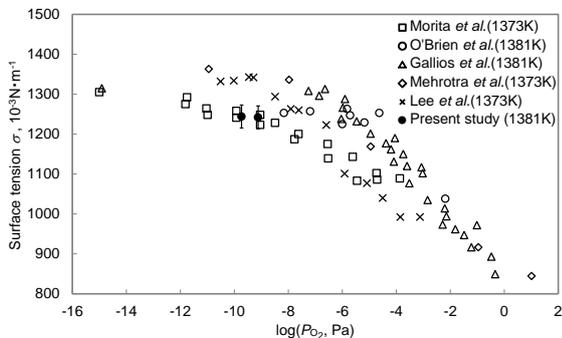


Fig. 2 Literature values of surface tension of molten copper as a function of oxygen partial pressure, P_{O_2} , of atmospheric gas at 1373K and 1381K together with our measurement data.

difficult to completely eliminate impurities such as aluminum and silicon from the sample. Thus, it is reasonable to suppose that the P_{O_2} should be decreased as low as possible to suppress the formation of oxides of those impurities.

Although there are small differences in the measurement temperature and absolute value of surface tension in **Fig. 2**, the surface tension seems to become constant when the P_{O_2} is less than 10^{-10} Pa. This implies that oxygen adsorption becomes less effective at a P_{O_2} less than 10^{-10} Pa. The aim was to control P_{O_2} at less than 10^{-10} Pa as a tentative target in this study in order to eliminate the effect of oxygen adsorption. It should be noted here that this P_{O_2} condition of less than 10^{-10} Pa can also be applied at temperatures higher than 1373K and 1381K corresponding to **Fig. 2** because the oxygen adsorption reaction is exothermic. That is, the influence of P_{O_2} on surface tension becomes small at a higher temperature.

3. Experimental confirmation of non-combustibility of atmospheric gas stipulated in the Japanese Civil Aeronautics Act

As mentioned above, we aimed to control the P_{O_2} at less than 10^{-10} Pa for the surface tension measurement of molten copper. In order to decrease the P_{O_2} of atmospheric gas, the gas phase equilibrium such as $\text{H}_2\text{-H}_2\text{O}$ and CO-CO_2 mixtures (see reactions 12 and 13) is usually used^{11-15, 17} because it is very difficult to obtain such a very low P_{O_2} even in high-purity commercial inert gas. However an airplane is not permitted to carry carbon monoxide gas due to its strong toxicity according to the Japanese Civil Aeronautics Act (CAA) (Article 86, Prohibition on Carriage of Explosives etc.). The employment of high concentration hydrogen gas is also prohibited in an airplane by the CAA because of its high combustibility. (When a lower explosive limit of gases mixed with air is less than or equal to 13% at 293K under 101.3kPa of absolute gas pressure, it is defined as an inflammable gas in the CAA.) However if a concentration of hydrogen becomes small by mixing with an inert gas such as argon, it would not be defined as an inflammable gas.

The criterion for dangerous goods that are prohibited to carry into an airplane is specified by the Ministry of Land, Infrastructure, Transport and Tourism in a directive from the CAA. According to this directive, whether gases are "combustible" or "non-combustible" can be determined through the experiment or calculation adopted in ISO 10156/1990 that is prescribed by the International Civil Aviation Organization (ICAO). Besides this method, another experiment method is described concretely in the directive.

In order to employ the Ar-3vol.%H₂ gas defined as nonflammable industrial gas in ISO 10156/1990 in the parabolic flight experiment, the non-combustibility of the gas was experimentally confirmed according to the directive. The results are described in this section. The temperature dependence of P_{O_2} of this gas was calculated as shown in Fig. 3⁹⁾ from the method mentioned in the previous section. This confirms that

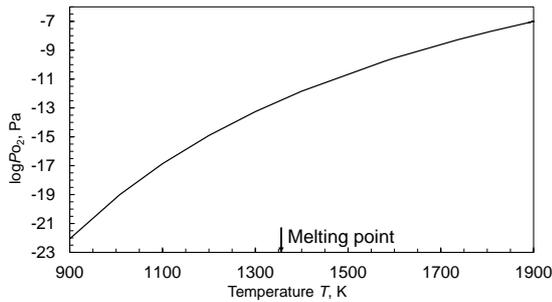


Fig. 3 Temperature dependence of oxygen partial pressure, P_{O_2} , of Ar-3 vol.%H₂ gas calculated from the standard Gibbs energy for formation of H₂O using the P_{O_2} measured by zirconia oxygen sensor operated at 1008K

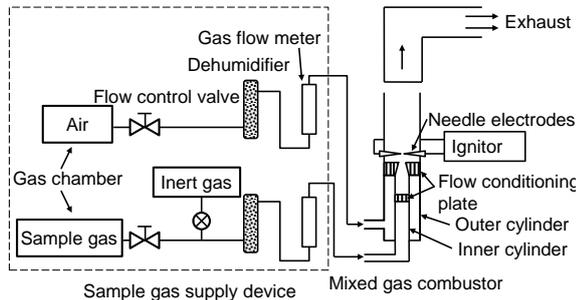


Fig. 4 Schematic of experimental setup for the gas combustion test stipulated in the civil aeronautics law.

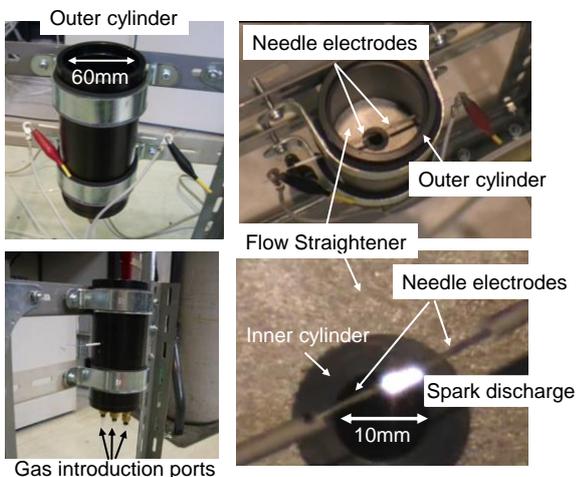


Fig. 5 Appearance of mixed gas combustor.

the P_{O_2} of the gas becomes less than 10⁻¹⁰ Pa below 1550K.

Figure 4 shows a schematic diagram of the experimental setup of the combustion test for high pressure gases stipulated in the CAA directive. The experimental apparatus consists of the mixed gas combustor (Fig. 5), gas supply unit, and ignitor. The mixed gas combustor is a double cylinder, in which the sample gas and the air are made to flow through the inner cylinder and outer cylinder, respectively. The inner cylinder has to have an exhaust nozzle of which the internal diameter of its top is 10 mm. The internal diameter of outer cylinder is specified to be 60 mm. Furthermore, a flow straightener for the gases must be mounted inside the cylinders.

The experimental method stipulated in the CAA directive is as follows:

- (1) The pipe for supplying the sample gas into the mixed gas combustor is evacuated and then backfilled by inert argon gas.
- (2) The sample gas flows into the combustor together with the air, in which the flow rate of the gases is precisely controlled at 1 cm/s at the outlet of the exhaust nozzle.
- (3) If spontaneous combustion of the gas is induced by the exposure of the gas to the air, the subsequent procedures should not be carried out.
- (4) After the streams of the gas and air become stabilized, an electric spark of discharge energy 20 mJ is generated between the needle electrodes positioned at 20 mm from the top center of the nozzle. The existence and non-existence of the gas ignition is visually judged, with gas ignition defined in this case as the situation whereby the flame is stabilized on the nozzle.
- (5) When gas ignition is not observed in three repetitions of the same procedure, the above procedures are performed at a sample gas flow rate of 3 cm/s and 5 cm/s while maintaining the flow rate of the air at 1 cm/s.
- (6) Moreover, if no gas ignition is confirmed even in procedure (5), procedures (1) to (5) are carried out again after the distance of the needle electrodes from the nozzle is reduced to 10 mm.
- (7) If the ignition of the gas is induced even once by the above procedures, the gas is defined as combustible.

From this experiment, it was confirmed that the Ar-3vol.%H₂ gas is a noncombustible gas, because ignition was never observed in the above experiments. Therefore, the gas is not dangerous goods as specified by the CAA.

4. Surface tension measurement of molten copper under a reducing gas atmosphere in the parabolic flight experiment

As mentioned above, it was confirmed that the Ar-3% H₂ gas is not dangerous goods by the experiment stipulated by the CAA.

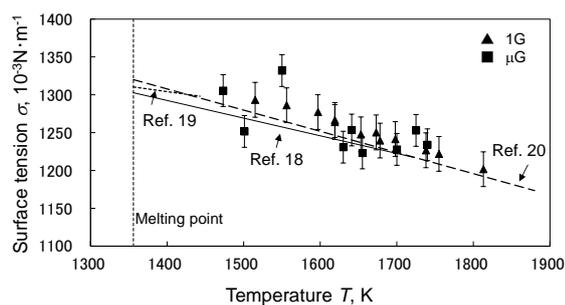


Fig. 6 The surface tension of molten copper calculated from the frequencies of $m=0, \pm 1$, and ± 2 oscillations under 1G (\blacktriangle) using Cumming and Blackburn calibration and that calibrated from the single frequency of the surface oscillation observed under microgravity (\blacksquare). Literature values are also shown¹⁸⁻²⁰⁾.

The surface tension of molten copper was measured by EML under the Ar-3vol.%H₂ gas in the microgravity condition during parabolic flight experiment⁹⁾. The results are shown in **Fig. 6** (\blacksquare) together with those measured in the terrestrial experiment (\blacktriangle). The literature data¹⁸⁻²⁰⁾ is also shown. The uncertainties for our measurement plot were calculated based on the GUM (ISO Guide to the Expression of Uncertainty in Measurement)²¹⁾, in which the coverage factor of $k_p=2$ was selected. The details of the experimental procedure and facilities can be found in the authors' earlier work⁹⁾.

The surface tension of molten copper was measured in the temperature range of 1470 K and 1820 K in the terrestrial experiment (\blacktriangle), which showed a comparatively good linear relationship against temperature. This agrees with the literature results. However it must be noted that there is still room for argument about whether the measurement results show the pure state of surface tension or not. Although it is implied that the P_{O_2} becomes less effective on the surface tension of molten copper at 1371 and 1381K when it is less than 10^{-10} Pa at 1371 K and 1381 K (see **Fig. 2**), the estimated P_{O_2} of the Ar-3vol.%H₂ gas used in this measurement exceeds 10^{-10} Pa above 1550 K as shown in **Fig. 3**.

We successfully measured the surface tension of molten copper under a reducing atmosphere of H₂-containing gas in a microgravity condition prepared by the parabolic flight of an airplane for the first time⁹⁾. The measurement result comparatively agrees with that of the terrestrial experiment.

However, it shows a small scattering compared with that measured on the ground. Two reasons for this scattering were suggested in our previous work⁹⁾. The first one is an incorrect estimation of the sample mass required for the surface tension calculation using the Rayleigh equation. Since the solidification of the sample was not finished within the short microgravity period of the parabolic flight experiment, the sample was not collected completely and the droplet fell at 1.5 G after the

microgravity condition. Accordingly, the variation of the sample mass during microgravity experiment was estimated from the initial sample mass and the results of the terrestrial experiment. The second possibility is that each calculated surface tension includes the influence of the temperature variation during the measurement. Since the electromagnetic force was significantly decreased under the microgravity condition to eliminate the influence of droplet deformation on surface oscillation, the temperature of droplet inevitably decreased by about 9K during the measurement.

In addition to these reasons, we would like to focus attention on the possibility of influence of P_{O_2} prepared by Ar-3vol.%H₂ gas in this manuscript. In our parabolic flight experiment, the P_{O_2} of the Ar-3vol.%H₂ gas was passively decided through the gas phase equilibrium described in reaction (12) depending on the amount of H₂O contained as impurity because it was not actively mixed into the gas. The volume content of H₂O existing as an impurity in the gas was calculated to be about 0.024 % from the standard Gibbs energy for formation of H₂O (eq. 15) using the P_{O_2} value of 10^{-19} Pa measured by the oxygen sensor operated at 1008K. This calculation method is the same as that for the temperature dependence of P_{O_2} corresponding to **Fig 3**.

Since the H₂O content in the gas was originally very little in this experiment, the ratio between the P_{H_2O} and P_{H_2} is sensitively changed by even an extremely small variation in the quantity of H₂O. It becomes difficult to assume that the ratio is constant regardless of temperature in this case. As a result, the P_{O_2} of the gas is also strongly influenced through reaction (12). For example, H₂O is dissociated corresponding to the temperature due to the temperature dependence of equilibrium constant K_{H_2O} (see eq. (14)). Furthermore, the distance from the gas supply system mounted on the airplane to our experimental facility is much longer than that for in the terrestrial experiment. In addition the gas supply system mounted on the airplane is used for various occasions between many users. Thus, it is necessary to pay full attention to the influence of physically adsorbed moisture inside the long pipes which carry the atmospheric gas during the measurement of surface tension.

Furthermore Kakikura et al.²²⁾ suggested that the P_{O_2} at the droplet surface is influenced by gas flow conditions such as inlet velocity and flow direction when the gravity level is significantly changed during the parabolic flight experiment.

In order to eliminate the influence of the variation of P_{O_2} due to an extremely small variation in the quantity of H₂O in the parabolic flight experiment, we suggest that moisture should be actively mixed into the Ar-3vol.%H₂ gas using a vaporizer in

future studies. For example, if the volume fraction of H₂O is increased from 0.024% to 1% in the gas, the P_{O_2} of the gas is not influenced by a small amount of dissociation of H₂O at high temperature because the variation of the ratio between P_{H_2O} and P_{H_2} becomes negligibly small due to the small variation in the quantity of the H₂O. It is also acceptable for a desorbed H₂O from the gas pipes.

Although the P_{O_2} of the Ar-3vol.%H₂ gas inevitably increased due to the gas phase equilibrium of the reaction (12) as shown in **Fig. 7** when the moisture content is increased, the P_{O_2} value can be selected by controlling the amount of mixed moisture according to the temperature. As a result, the surface tension measurement at an isothermal condition and iso- P_{O_2} condition can be expected under a microgravity condition as shown by the dashed line and dotted line, respectively.

5. Summary

The surface tension measurements of molten metal using the EML under microgravity condition during the parabolic flight of an airplane were described in this manuscript. Copper was selected as the sample for the parabolic flight experiment from the viewpoint of its oxidization tendency, the influence of oxygen partial pressure, and ease of melting and electromagnetic levitation. Since it was confirmed that the Ar-3vol.%H₂ gas is not dangerous by the experiment stipulated in the Civil Aeronautics Act, it was employed in the parabolic flight experiment to decrease the P_{O_2} . Although the surface tension of molten copper measured under microgravity condition was similar to that measured on the ground, it showed a small scattering due to the incorrect estimation of the sample mass and temperature variation during the measurement. We also pointed out that the P_{O_2} of the Ar-3vol.%H₂ gas is influenced by even an extremely small variation in the quantity of H₂O. Furthermore, we suggested that moisture should be actively mixed into the Ar-3vol.%H₂ gas to decrease a large sensitivity of the P_{O_2} of the gas to an extremely small variation in the quantity of H₂O.

Acknowledgement

This research was funded by the JAXA Research WG. This work was partially supported by JSPS KAKENHI Grant Number 24760617 and the Sasakawa Scientific Research Grant

from The Japan Science Society. The authors thank Mr. D. Kageyama and the other staff of Diamond Air Service, Inc. for their assistance in the parabolic flight experiment. We also thank Prof. S. Yuasa (Tokyo Metropolitan Univ.), Dr. T. Sakurai, (Tokyo Metropolitan Univ.) and Prof. O. Fujita (Hokkaido Univ.) for their helpful comments on the safety of the combustion of atmosphere gas.

References

- 1) I. Egry, E. Ricci, R. Novakovic and S. Ozawa: *Adv. Colloid Interface Sci.*, **159** (2010) 198.
- 2) S. Ozawa, T. Koda, M. Adachi, K. Morohoshi, M. Watanabe and T. Hibiya: *J. Appl. Phys.*, **106** (2009) 034907-1.
- 3) S. Ozawa, K. Morohoshi, T. Hibiya and H. Fukuyama: *J. Appl. Phys.*, **107** (2010) 014910-1.
- 4) Lord Rayleigh: *Proceedings of the Royal Society of London*, **29** (1879) 71.
- 5) D. L. Cummings and D. A. Blackburn: *J. Fluid Mech.*, **224** (1991) 395.
- 6) I. Egry, G. Lohoefer and G. Jacobs: *Physical Review Letters*, **75** (1995) 4043.
- 7) I. Egry, G. Lohöfer and D. Matson: *High Temp. Mater. Process.*, **27** (2008) 389.
- 8) H. Fujii, T. Matsumoto, and K. Nogi: *Acta. mater.*, **48** (2000) 2933.
- 9) S. Ozawa, M. Watanabe, Y. Kiyamura, K. Morohoshi, T. Aoyagi, M. Tanno, T. Matsumoto, M. Adachi, A. Mizuno, H. Fujii and T. Hibiya: *J. Jpn. Soc. Microgravity Appl.*, **27** (2010) 215.
- 10) M. W. Chase, Jr.: *NIST-JANAF Thermochemical Tables*, 4th ed., The American Chemical Society and the American Institute of Physics for the National Institute of Standards and Technology, New York, 1998.
- 11) Z. Morita and A. Kasama: *J. Jpn. Inst. of Metal*, **16** (1975) 787 (in Japanese).
- 12) T. E. O'Brien and A. C. D. Chaklader: *J. Am. Ceram. Soc.*, **57** (1974) 329.
- 13) B. Gallios and C. H. P. Lupis: *Metall. Trans.*, **B12** (1981) 549.
- 14) S. P. Mehrotra and A. C. D. Chaklader: *Metall. Trans.*, **B16** (1985) 567.
- 15) J. Lee, T. Tanaka, Y. Asano and S. Hara: *Materials transactions*, **45** (2004) 2719.
- 16) O. Knacke, O. Kubaschewski, K. Hesselmann: *Thermochemical properties of inorganic substances*, 2nd ed., p. 811, Springer-Verlag, Verlag Stahleisen, 1991.
- 17) S. Ozawa, S. Takahashi and Y. Takei: *Proceeding of Interfacial Joining and Surface Technology*, 2013.
- 18) A. Kasama, T. Iida, and Z. Morita: *J. Jpn. Inst. of Metals*, **16** (1976) 787. (in Japanese)
- 19) S. K. Rhee: *J. American Ceramic Soc.*, **53** (1970) 639.
- 20) K. Nogi, K. Oishi and K. Ogino: *J. Jpn. Inst. of Metals*, **52** (1988) 72. (in Japanese)
- 21) *Guide to the Expression of Uncertainty in Measurement*, ISO, 1995.
- 22) K. Kakikura, K. Fukagata, and T. Hibiya: *J. Jpn Soc. Microgravity Appl.*, **28** (2011) S92.

(Received 14 Nov. 2013; Accepted 17 Jan. 2014)